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| FORM PTO-1390 (REV. 11-2000) | | U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE | | ATTORNEY'S DOCKET NUMBER A01058US (98148.13) | |
| TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371 | | | | U.S. APPLICATION NO. (If known, see 37 CFR 1.5) | |
| | | | | 09/786719 | |
| INTERNATIONAL APPLICATION NO. PCT/RU99/00273 | | INTERNATIONAL FILING DATE 2 August 1999 (02.08.99) | | PRIORITY DATE CLAIMED 8 September 1998 (08.09.98) | |
| TITLE OF INVENTION "Combustion Retardant for Polymeric Materials" | | | | | |
| APPLICANT(S) FOR DO/EO/US | | | | | |
| Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information: | | | | | |
| <ol style="list-style-type: none"> 1. <input checked="" type="checkbox"/> This is a FIRST submission of items concerning a filing under 35 U.S.C. 371. 2. <input type="checkbox"/> This is a SECOND or SUBSEQUENT submission of items concerning a filing under 35 U.S.C. 371. 3. <input type="checkbox"/> This is an express request to begin national examination procedures (35 U.S.C. 371(f)). The submission must include items (5), (6), (9) and (21) indicated below. 4. <input type="checkbox"/> The US has been elected by the expiration of 19 months from the priority date (Article 31). 5. <input checked="" type="checkbox"/> A copy of the International Application as filed (35 U.S.C. 371(c)(2)) <ol style="list-style-type: none"> a. <input checked="" type="checkbox"/> is attached hereto (required only if not communicated by the International Bureau). b. <input type="checkbox"/> has been communicated by the International Bureau. c. <input type="checkbox"/> is not required, as the application was filed in the United States Receiving Office (RO/US). 6. <input checked="" type="checkbox"/> An English language translation of the International Application as filed (35 U.S.C. 371(c)(2)). <ol style="list-style-type: none"> a. <input checked="" type="checkbox"/> is attached hereto. b. <input type="checkbox"/> has been previously submitted under 35 U.S.C. 154(d)(4). 7. <input checked="" type="checkbox"/> Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3)) <ol style="list-style-type: none"> a. <input type="checkbox"/> are attached hereto (required only if not communicated by the International Bureau). b. <input type="checkbox"/> have been communicated by the International Bureau. c. <input type="checkbox"/> have not been made; however, the time limit for making such amendments has NOT expired. d. <input checked="" type="checkbox"/> have not been made and will not be made. 8. <input type="checkbox"/> An English language translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371 (c)(3)). 9. <input type="checkbox"/> An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)). 10. <input type="checkbox"/> An English language translation of the annexes of the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)). | | | | | |
| <p>▪ Items 11 to 20 below concern document(s) or information included:</p> <ol style="list-style-type: none"> 11. <input type="checkbox"/> An Information Disclosure Statement under 37 CFR 1.97 and 1.98. 12. <input type="checkbox"/> An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included. 13. <input type="checkbox"/> A FIRST preliminary amendment. 14. <input type="checkbox"/> A SECOND or SUBSEQUENT preliminary amendment. 15. <input type="checkbox"/> A substitute specification. 16. <input type="checkbox"/> A change of power of attorney and/or address letter. 17. <input type="checkbox"/> A computer-readable form of the sequence listing in accordance with PCT Rule 13ter.2 and 35 U.S.C. 1.821 - 1.825. 18. <input checked="" type="checkbox"/> A second copy of the published international application under 35 U.S.C. 154(d)(4). 19. <input checked="" type="checkbox"/> A second copy of the English language translation of the international application under 35 U.S.C. 154(d)(4). 20. <input type="checkbox"/> Other items or information: | | | | | |



09786719-00273

U.S. APPLICATION NO. (if known, see 37 CFR 1.53)
097786719

INTERNATIONAL APPLICATION NO.

ATTORNEY'S DOCKET NUMBER

21. ☒ The following fees are submitted:

BASIC NATIONAL FEE (37 CFR 1.492 (a) (1) - (5)):

Neither international preliminary examination fee (37 CFR 1.482)
nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO
and International Search Report not prepared by the EPO or JPO..... **\$1000.00**

International preliminary examination fee (37 CFR 1.482) not paid to
USPTO but International Search Report prepared by the EPO or JPO **\$860.00**

International preliminary examination fee (37 CFR 1.482) not paid to USPTO
but international search fee (37 CFR 1.445(a)(2)) paid to USPTO **\$710.00**

International preliminary examination fee (37 CFR 1.482) paid to USPTO
but all claims did not satisfy provisions of PCT Article 33(1)-(4) **\$690.00**

International preliminary examination fee (37 CFR 1.482) paid to USPTO
and all claims satisfied provisions of PCT Article 33(1)-(4) **\$100.00**

ENTER APPROPRIATE BASIC FEE AMOUNT =

CALCULATIONS PTO USE ONLY

\$ 1,000

Surcharge of **\$130.00** for furnishing the oath or declaration later than ☐ 20 ☒ 30
months from the earliest claimed priority date (37 CFR 1.492(e)).

\$ 130

| CLAIMS | NUMBER FILED | NUMBER EXTRA | RATE | \$ |
|---|--------------|--------------|-------------------|----|
| Total claims | - 20 = | | x \$18.00 | \$ |
| Independent claims | - 3 = | | x \$80.00 | \$ |
| MULTIPLE DEPENDENT CLAIM(S) (if applicable) | | | + \$270.00 | \$ |

TOTAL OF ABOVE CALCULATIONS = \$ 1,130

☒ Applicant claims small entity status. See 37 CFR 1.27. The fees indicated above
are reduced by 1/2.

\$ 565

SUBTOTAL = \$ 565

Processing fee of **\$130.00** for furnishing the English translation later than ☐ 20 ☐ 30
months from the earliest claimed priority date (37 CFR 1.492(f)).

\$

TOTAL NATIONAL FEE = \$ 565

Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be
accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). **\$40.00** per property +

\$

TOTAL FEES ENCLOSED = \$ 565

**Amount to be
refunded:**

\$

charged:

\$

- a. ☒ A check in the amount of **\$ 565.00** to cover the above fees is enclosed.
- b. ☐ Please charge my Deposit Account No. _____ in the amount of \$ _____ to cover the above fees.
A duplicate copy of this sheet is enclosed.
- c. ☒ The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any
overpayment to Deposit Account No. **50-0694**. A duplicate copy of this sheet is enclosed.
- d. ☐ Fees are to be charged to a credit card. **WARNING:** Information on this form may become public. **Credit card
information should not be included on this form.** Provide credit card information and authorization on PTO-2038.

NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR
1.137 (a) or (b)) must be filed and granted to restore the application to pending status.

SEND ALL CORRESPONDENCE TO:

Seth M. Nehrbass

PTO CUSTOMER NUMBER 22920

SIGNATURE

Seth M. Nehrbass

NAME

31,281

REGISTRATION NUMBER



J004/Rec'd PCT/PTO 22 JUN 2001

APPLICANTS: Zubkova et al

DATE: June 18, 2001

U.S. SERIAL NO.: 09/786,719

GROUP ART UNIT:

INTERNATIONAL FILING DATE: 02 August 1999

EXAMINER:

FOR: "COMBUSTION RETARDANT FOR POLYMERIC MATERIALS"

ATTORNEY DOCKET NO.: A01058US (98148.13)

**PRELIMINARY AMENDMENT AND
RESPONSE TO NOTICE TO FILE MISSING PARTS**

Commissioner of Patents
and Trademarks
Washington, D.C. 20231

Sir:

This is an amendment and response to the notice mailed from the U.S. Patent Office on
17 April 2001 in the above-referenced application. The deadline to respond to this notice is 18
June 2001 as 17 June 2001 was a holiday.

AMENDMENT

Please amend the application as follows:

Adjustment date: 08/05/2001 WDI AYSRQ
07/10/2001 WDI AYSRQ
01 FC:967 -252.00 DP
02 FC:998 -3.00 DP

~~IN THE CLAIMS~~

Please amend the claims to read as follows:

--1. Complex compound of ammonia salt of amide of alkylphosphonic acid with

~~ammonium chloride of formula (I)~~

Repln. Ref: 08/05/2001 WDI AYSRQ
DRI:000694
FC: 704

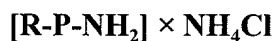
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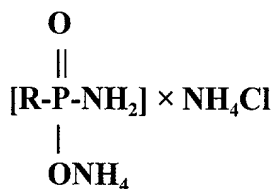


where R is the alkyl radical C-1-3.

2. Complex compound in accordance with Claim 1, in which there are about 1.8 molecules of ammonium chloride to one molecule of ammonia salt of amide of alkylphosphonic acid.

3. Process for producing a complex compound of ammonia salt of amide of alkylphosphonic acid with ammonium chloride of Formula (I), consisting of the interaction of dichloroanhydride of alkylphosphonic acid with gaseous ammonia in a medium of organic solvent at temperature 10-20°C.

4. Combustion retardant for polymer materials, consisting of a complex compound of ammonia salt of amide of alkylphosphonic acid with ammonium chloride of formula (I)



where R is the alkyl radical C-1-3.

5. Combustion retardant in accordance with Claim 4, in which there are about 1.8 molecules of ammonium chloride to one molecule of ammonia salt of amide of alkylphosphonic acid.

6. (Amended) Combustion retardant in accordance with Claim 4, *characterised* in that it is microencapsulated in a polymer shell.

7. Combustion retardant in accordance with Claim 6, *characterised* in that the said

polymer shell is made of polyethylene.

8. Combustion retardant in accordance with Claim 6, *characterised* in that the said polymer shell is made of polyorganosiloxanes.

9. Combustion retardant in accordance with Claim 8, *characterised* in that the polyorganosiloxanes are selected from a group including polyvinylmethyldiethoxy-siloxane and polyaminopropylethoxysiloxane.

10. Process for producing low fire risk polymer materials by the introduction of the CR into the polymer in the course of its processing, *characterised* in that the CR used is a complex compound of ammonia salt of amide of alkylphosphonic acid with ammonium chloride of Formula (I).

11. Process for producing low fire risk polymer materials in accordance with Claim 10, *characterised* in that it includes the following sequence of operations:

- joint extrusion of the said combustion retardant with the polymer;
- moulding the polymer fibre;
- granulation.

12. Process for producing low fire risk polymer materials in accordance with Claim 10, *characterised* in that it includes the following sequence of operations:

- mixing of the said combustion retardant with the polymer composition;
- rolling the mass;
- pressing the articles.

13. (Amended) Process in accordance with Claim 10, *characterised* in that the combustion retardant is first microencapsulated in a polymer shell.

14. Process in accordance with Claim 13, *characterised* in that the size of the

microcapsules is 5-25 $\frac{1}{4}$ m.

15. Process in accordance with Claim 13, *characterised* in that the polymer shell is made of polyethylene with shell content including 10-15 wt.% of combustion retardant.

16. Process in accordance with Claim 13, *characterised* in that polyorganosiloxanes are used for the polymer shell.

17. Process in accordance with Claim 16, *characterised* in that the polyorganosiloxane consists of polymethyldiethoxysiloxanes with shell containing 2-5 wt.% of combustion retardant.

18. Process in accordance with Claim 16, *characterised* in that the polyorganosiloxane used is polyaminopropylethoxysiloxane, with shell containing 2-5 wt.% of combustion retardant.

19. (Amended) Process in accordance with Claim 10, *characterised* in that polyethylene, polypropylene and copolymers of various compositions based on them are processed.

20. (Amended) Process in accordance with Claim 10, *characterised* in that polystyrene and copolymers of various compositions based on it are processed.

21. Process for producing low fire risk polymer materials by the introduction of combustion retardant into the polymer, *characterised* in that the combustion retardant used is a complex compound of ammonia salt of amide of alkylphosphonic acid with ammonium chloride of Formula (I), which is introduced into the polymer composition before it sets.

22. Process in accordance with Claim 21, *characterised* in that a filler is introduced into the polymer composition along with the said combustion retardant, and as a result of the saturation of the filler with the setting polymer composition, low fire risk materials are produced.

23.(Amended) Process in accordance with Claim 21, *characterised* in that polyesters are

processed.

24. (Amended) Process in accordance with Claim 21, *characterised* in that epoxy resins are processed.

25. Process for producing low fire risk polymer materials, *characterised* in that a complex compound of ammonia salt of amide of alkylphosphonic acid with ammonium chloride of Formula (I) is introduced into a polymer composition including synthetic rubber, after which it is rolled before the article is pressed.

26. Low fire risk polyethylene produced by a process in accordance with Claim 6.

27. Low fire risk polypropylene produced by a process in accordance with Claim 6.

28. Low fire risk polystyrene produced by a process in accordance with Claim 6.

29. Low fire risk copolymers based on polyethylene, polypropylene and polystyrene, produced by a process in accordance with Claim 6.

30. Low fire risk polyesters produced by a process in accordance with Claim 21.

31. Low fire risk epoxy resins produced by a process in accordance with Claim 21.

32. Low fire risk composition materials produced by a process in accordance with Claim 22.

33. Low fire risk synthetic rubbers produced by a process in accordance with Claim 25.

34. Low fire risk polycaproamide materials produced by a process in accordance with Claim 13.

35. Low fire risk polymethyl methacrylate compositions produced by a process in accordance with Claim 11.--

REMARKS

The claims have been amended to reduce claims fees. For convenience in prosecution, all claims are repeated herein, even those which are not amended herein. Two sets of claims are included, one set (attached) showing the changes made in this response and one clean set (set out above). No new matter has been added to the application.

Enclosed are the following:

1. Declaration;
2. Power Of Attorney by the Assignee;
3. Statement under 37 C.F.R. § 3.73(b);
4. Copy of Assignment (The assignment has been requested to be recorded with the Patent and Trademark Office);
5. \$255.00 for Excess Claim Fees (Applicant qualifies for small entity status); and
6. Copy of Formalities letter;

Applicants have not included the Oath Surcharge under 37 C.F.R. §1.492(e) because this surcharge was paid with the original papers entering the national stage. If this is incorrect, the office is authorized to charge said surcharge to Deposit Account Number 50-0694.

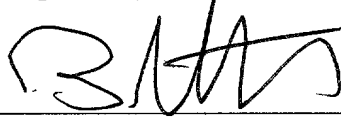
Applicants respectfully submit that the application is in condition for allowance. A Notice of Allowance is hereby respectfully requested.

Should the Examiner feel that a telephone conference would advance the prosecution of this application, he is encouraged to contact the undersigned at the telephone number listed below.

Applicants respectfully petition the Commissioner for any extension of time necessary to render this paper timely.

Please charge any fees due or credit any overpayment to Deposit Account No. 50-0694.

Respectfully submitted,



Seth M. Nehrbass, Reg. No. 31,281

Charles C. Garvey, Jr., Reg. No. 27,889

Gregory C. Smith, Reg. No. 29,441

Stephen R. Doody, Reg. No. 29,062

H. Roy Berkenstock, Reg. No. 24,719

Brett A. North, Reg. No. 42,040

GARVEY, SMITH, NEHRBASS & DOODY, L.L.C.

PTO Customer No.: 22920

3838 N. Causeway Blvd., Suite 3290

Metairie, LA 70002

Tel.: (504) 835-2000

Fax: 504-835-2070

e-mail: IPLNO@AOL.COM

www.neworleanspatents.com

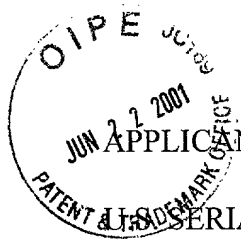
CERTIFICATE OF MAILING

I hereby certify that this correspondence is being deposited with the United States Postal Service as First Class Mail in an envelope addressed to: Commissioner of Patents and Trademarks, Washington, D.C. 20231, on this 18 day of June, 2001.



Brett A. North, Reg. No. 31,281

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APPLICANTS: Zubkova et al

DATE: June 18, 2001

SERIAL NO.: 09/786,719

GROUP ART UNIT:

INTERNATIONAL FILING DATE: 02 August 1999

EXAMINER:

FOR: "COMBUSTION RETARDANT FOR POLYMERIC MATERIALS"

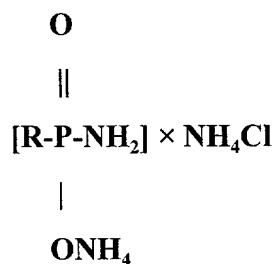
ATTORNEY DOCKET NO.: A01058US (98148.13)

* * * * *

Copy of claims showing changes

Set out below is a copy of claims showing changes made in this amendment, with added matter underlined and omitted matter in brackets:

-- 1. Complex compound of ammonia salt of amide of alkylphosphonic acid with ammonium chloride of formula (I)

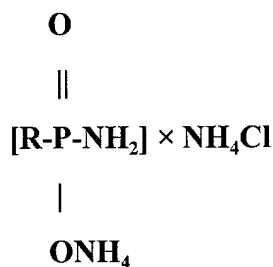


where R is the alkyl radical C-1-3.

2. Complex compound in accordance with Claim 1, in which there are about 1.8 molecules of ammonium chloride to one molecule of ammonia salt of amide of alkylphosphonic acid.

3. Process for producing a complex compound of ammonia salt of amide of alkylphosphonic acid with ammonium chloride of Formula (I), consisting of the interaction of dichloroanhydride of alkylphosphonic acid with gaseous ammonia in a medium of organic solvent at temperature 10-20°C.

4. Combustion retardant for polymer materials, consisting of a complex compound of ammonia salt of amide of alkylphosphonic acid with ammonium chloride of formula (I)



where R is the alkyl radical C-1-3.

5. Combustion retardant in accordance with Claim 4, in which there are about 1.8 molecules of ammonium chloride to one molecule of ammonia salt of amide of alkylphosphonic acid.

6. (Amended) Combustion retardant in accordance with Claim 4 [or 5], *characterised* in that it is microencapsulated in a polymer shell.

7. Combustion retardant in accordance with Claim 6, *characterised* in that the said polymer shell is made of polyethylene.

8. Combustion retardant in accordance with Claim 6, *characterised* in that the said polymer shell is made of polyorganosiloxanes.

9. Combustion retardant in accordance with Claim 8, *characterised* in that the polyorganosiloxanes are selected from a group including polyvinylmethyldiethoxy-siloxane and polyaminopropylethoxysiloxane.

10. Process for producing low fire risk polymer materials by the introduction of the CR into the polymer in the course of its processing, *characterised* in that the CR used is a complex compound of ammonia salt of amide of alkylphosphonic acid with ammonium chloride of Formula (I).

11. Process for producing low fire risk polymer materials in accordance with Claim 10, *characterised* in that it includes the following sequence of operations:

- joint extrusion of the said combustion retardant with the polymer;
- moulding the polymer fibre;
- granulation.

12. Process for producing low fire risk polymer materials in accordance with Claim 10, *characterised* in that it includes the following sequence of operations:

- mixing of the said combustion retardant with the polymer composition;
- rolling the mass;
- pressing the articles.

13. (Amended) Process in accordance with [any of] Claim[s] 10[-12], *characterised* in that the combustion retardant is first microencapsulated in a polymer shell.

14. Process in accordance with Claim 13, *characterised* in that the size of the microcapsules is 5-25 μm .

15. Process in accordance with Claim 13, *characterised* in that the polymer shell is made of polyethylene with shell content including 10-15 wt.% of combustion retardant.

16. Process in accordance with Claim 13, *characterised* in that polyorganosiloxanes are used for the polymer shell.

17. Process in accordance with Claim 16, *characterised* in that the polyorganosiloxane consists of polymethyldiethoxysiloxanes with shell containing 2-5 wt.% of combustion retardant.

18. Process in accordance with Claim 16, *characterised* in that the polyorganosiloxane used is polyaminopropylethoxysiloxane, with shell containing 2-5 wt.% of combustion retardant.

19. (Amended) Process in accordance with [any of] Claim[s] 10[-18], *characterised* in that polyethylene, polypropylene and copolymers of various compositions based on them are processed.

20. (Amended) Process in accordance with [any of] Claim[s] 10[-18], *characterised* in that polystyrene and copolymers of various compositions based on it are processed.

21. Process for producing low fire risk polymer materials by the introduction of combustion retardant into the polymer, *characterised* in that the combustion retardant used is a complex compound of ammonia salt of amide of alkylphosphonic acid with ammonium chloride of Formula (I), which is introduced into the polymer composition before it sets.

22. Process in accordance with Claim 21, *characterised* in that a filler is introduced into the polymer composition along with the said combustion retardant, and as a result of the saturation of the filler with the setting polymer composition, low fire risk materials are produced.

23.(Amended) Process in accordance with Claim 21[or 22], *characterised* in that polyesters are processed.

24. (Amended) Process in accordance with Claim 21[or 22], *characterised* in that epoxy resins are processed.

25. Process for producing low fire risk polymer materials, *characterised* in that a complex compound of ammonia salt of amide of alkylphosphonic acid with ammonium chloride of Formula (I) is introduced into a polymer composition including synthetic rubber, after which it is rolled before the article is pressed.

26. Low fire risk polyethylene produced by a process in accordance with Claim 6.

27. Low fire risk polypropylene produced by a process in accordance with Claim 6.

28. Low fire risk polystyrene produced by a process in accordance with Claim 6.

29. Low fire risk copolymers based on polyethylene, polypropylene and polystyrene,

produced by a process in accordance with Claim 6.

30. Low fire risk polyesters produced by a process in accordance with Claim 21.

31. Low fire risk epoxy resins produced by a process in accordance with Claim 21.

32. Low fire risk composition materials produced by a process in accordance with Claim 22.

33. Low fire risk synthetic rubbers produced by a process in accordance with Claim 25.

34. Low fire risk polycaproamide materials produced by a process in accordance with Claim 13.

35. Low fire risk polymethyl methacrylate compositions produced by a process in accordance with Claim 11. --

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COMBUSTION RETARDANT FOR POLYMERIC MATERIALS**Technology field**

5 The invention relates to a technology for producing polymer compositions based on carbon-chain polymers (polyethylene, polypropylene, polystyrene, synthetic rubbers and copolymers of various compositions), heterochain polymers (polyester, epoxy and phenol resins) and composition materials of various compositions and fillings with low combustibility, low toxicity of gases emitted during combustion and
10 low smoke emission.

Polymer materials are widely used in the cable and motor industries, electrical consumer goods, construction, other consumer goods, the gas and oil extraction industries, aviation and space technology and for making packing materials.

15 Prior Art

A significant problem with the majority of industrially produced polymer materials is their high flammability and high rate of combustion, accompanied by the emission of a large quantity of toxic products.

20 With the aim of reducing the combustibility of carbon-chain polymers, physical (Kistelman V.I. Physical Methods of Modifying Polymer Materials, Moscow, Khimiya, 1980, 223 pp.) and chemical methods of modification are used, and also a combination of them, e.g. photochemical modification (Kachan A.A., Zamotayev P.V. The Photochemical Modification of Polyolefins, Kiev, Naukova
25 dumka, 1990, 276 p.). Chemical modification by means of halogenation brings about greater degree of reduction in their combustibility. However, to use this method to obtain a polyolefin which is extinguished when the external heat source is removed, it is necessary to chlorinate polyethylene (PE) and polypropylene (PP) to a halogen content of 25-40 wt.% (Aseyeva R.M., Zaikov G.Ye. The Combustion of Polymer
30 Materials, Moscow, Nauka, 1991, 150 pp.). With such a chlorine content, the crystallinity of PE and PP is sharply reduced, so that they are transformed from

thermoplastic into elastomers (Sirota A.G. Modification of the Structure and Properties of Polyolefins, Moscow, Khimiya, 1984, 150 pp.). Chlorinated PE finds application as a low combustibility material in its own right and as a combustion retardant (CR) of a polymer nature for other polymer materials. The main problems with chlorinated polyolefins are their low thermal stability and their emission of toxic products, which limit their application.

Polymers with higher thermal stability and oxygen index (OI) (above 27%) can be obtained by sulfochlorination (Aseyeva R.M., Zaikov G.Ye. The Combustion of Polymer Materials, Moscow, Nauka, 1991, 150 pp.). Like chlorination, sulfochlorination can lead to the formation of elastomers.

(N.B. OI, the oxygen index, is the minimum content of oxygen in a mixture with nitrogen at which stable combustion of a specimen is observed.)

For the chemical modification of polystyrene, styrene is copolymerised with monomers containing chlorine, bromine or phosphorus: vinyl chloride, vinyl bromide, vinylidene chloride, chlorinated and bromated styrenes, acrylates containing halogens, halogenated fumarates, N-phenylmaleimides, phosphorylated styrene, halogenated esters of vinyl- and allylphosphonic acids, phenyldichlorophosphine and tris(methacryloylbromomethyl) phosphate (Low Combustibility Polymer Materials, ed. A.N. Pravednikov, Moscow, Khimiya, 1986, 132 pp.).

The method of chemical modification of carbon-chain polymers with the aim of imparting fireproofing properties to them enables a fireproofing effect which is resistant to various treatments to be obtained. However, it requires changes in the technology for producing the polymer, and leads to the appearance of a number of negative properties in the end product, which limits the application possibilities for this method.

In scale of use, chemical modification methods lag far behind the method of introducing CRs and systems of them at the processing stage of the polymers (Berlin A.A., Volfson S.A., Oshmyan V.G. et al. Principles of the Creation of Fireproofed Polymer Materials, Moscow, Khimiya, 1990, 240 pp.)

The process of producing low combustibility synthetic materials by the introduction of CRs into the polymer melt during moulding makes it possible to retain the existing technology for processing articles, is highly economical and creates conditions for developing ecologically clean processes. It also ensures that the fireproofing is highly resistant to wet treatments.

As CRs for rubbers, the most widely used are aluminium trihydroxide and aluminium oxide, which not only reduce the combustibility of the rubber, but also eliminate the disadvantage of smoke emission.

However, to produce compositions which do not support combustion in air, the degree of filling of the polymer composition with combustion retardant must be not less than 50%, which complicates the process of treating the compositions and reduces the physical and mechanical indicators (Low Combustibility Polymer Materials, ed. A.N. Pravednikov, Moscow, Khimiya, 1986, 132 pp.).

There are known joint uses of $\text{Al}(\text{OH})_3$ and $\text{Mg}(\text{OH})_2$ in combination with swelling graphite (Khokhlova L.A., Aseyeva R.P., Ruban L.V., International Conference on Low Combustibility Polymer Materials. Alma-Ata, 1990, Vol. 1, pp. 16-18).

A big problem in processing inert CRs is the migration of additives (not compatible with the polymer lattice) from the polymer lattice to its surface, since these additives are not bonded to it. This leads to a reduction in the fire retardant effect, and in contact with the surface of metals, increases corrosion activity with the surface of the metals.

More effective CRs for polyolefins and synthetic rubbers are bromoorganic ones which are introduced into polymers in combination with a synergic additive – antimony trioxide (US 5116898, MPC C 08K 5/06). The replacement of part of the trioxide enables the CR content to be reduced. To reduce the combustibility of polystyrene, halogenated aliphatic compounds are used in combination with antimony trioxide: chloroparaffins, perchlorinated alkanes C_2Cl_6 , C_4Cl_{10} , aliphatic compounds containing bromine (tetrabromethane, tetrabromoctane, 1,2,3,4 – tetrabromine 2,3- dimethylbutane, 2,3,4,5 – tetrabromine-2,5-dimethylhexane and

others (Low Combustibility Polymer Materials, ed. A.N. Pravednikov, Moscow, Khimiya, 1986, 132 pp.).

To impart self-extinguishing properties to polyolefins and synthetic rubbers, organic CRs must be used in high concentrations (up to 40% chlorine or 20-30% bromine).

Several publications describe the use of red phosphorus (the polymer form of elemental phosphorus) as a CR for polyolefins (Low Combustibility Polymer Materials, ed. A.N. Pravednikov, Moscow, Khimiya, 1986, 132 pp.). A polyethylene with OI 26.2% has an 8% content of phosphorus. However, in processing polyolefins containing red phosphorus, toxic phosphorous hydrogen (phosphine) is emitted.

There is a known use of ammonium polyphosphates as CRs for polyolefins and synthetic rubbers (Application 2272444 Great Britain, MPC C 08F 8/40, C08F 9/44).

The effectiveness of the action of ammonium polyphosphates depends on how finely they are crushed. However, even at a fine degree of dispersion, a high degree of filling (40-50 wt.%) is required to achieve OI 28%, which leads to a considerable reduction in the physical and mechanical properties of the material.

Many studies have been devoted to the synthesis of amides or alkylamides of phosphoric or alkylphosphonic acid and their use as CRs to impart fireproofing properties to polymer materials. The studies carried out by Drews (Drews M.J., Textilveredlung, 1973, Vol. 8, pp 180-186) showed that compounds containing a P-N bond are more effective CRs than compounds with P-O bonds. The synthesis of phosphorus triamide has been described (Herlinger H. Textilveredlung, 1977, Vol. 12, pp. 13-20) and it is proposed that it should be used to impart fireproofing properties to cellulose materials. The reaction was conducted by the interaction of trichloroanhydride of phosphoric acid with ammonia in chloroform at temperature 10°C. A problem with the CR thus obtained is a reduction of the physical and mechanical indicators of polymer materials modified by this CR by 50-60%.

With the aim of eliminating this problem, pentamethylphosphorotriamide was synthesised by treating phosphorus oxychloride with dimethylamine and

methylamine (L Blanc R.B., Text. Chem. Colorist, 1975, Vol. 7, No. 10, pp. 23-25). However, the synthesised compounds possessed high thermal stability, and were therefore less effective as fireproofing for polymer materials.

A method of synthesising diamide of methylphosphonic acid by treating
5 dichloroanhydride of methylphosphonic acid with liquid ammonia in a chloroform medium was proposed in another work (Ratz R.J., Am. Chem. Soc., 1955, Vol. 77, pp. 4170-4171). All the reagents, including the solvent, were dehydrated. However, as was shown in this work, diamide of methylphosphonic acid, which is separated out from the reaction mixture by boiling in a medium of diethylamine and chloroform,
10 has low resistance to the effect of hydrolising agents, and even under the effect of the moisture in the air, diamide of methylphosphonic acid gradually passes through the ammonia salt into the methylphosphonic acid. Due to this problem, this compound cannot be recommended as a CR for introduction into molten polymer.

With the aim of eliminating this fault, RU Patent No. 20993384 proposed the
15 microencapsulation of partly hydrolised diamide of methylphosphonic acid – the ammonium salt of amide of methylphosphonic acid – in a heat-resistant shell based on polyaramides. However, the CR produced has an insufficiently effective fireproofing action for polyolefins, and can be recommended only for a reduction in the combustibility of polyamides and polyesters. It should be noted here that it is
20 difficult to conduct the process of microencapsulation into polyaramide shells without structural defects.

There is a known use of organosilicon compounds to modify CRs and to make it easier to process compositions with high degrees of filling. To make processing easier, modifying additives are introduced into the compositions: e.g.,
25 there is a known low-combustibility composition (Bolikhova V.D., Drobinin A.N. Plastic Masses, Moscow, Z.-S. 1994, pp. 46-51) including the antipyrene $\text{Al}(\text{OH})_3$, and as a modifying additive, silanic and polysiloxanic acids.

Organic compounds containing halogens are used to modify heterochain polymers, in particular polyesters. These are mainly aromatic CRs containing
30 bromine. They are used because of their higher thermal stability and lower smoke emission in comparison with aliphatic compounds containing halogens (Namets

R.C., *Plastics Compounding*, 1984, Vol. 7, No. 4, pp. 26-39). To reduce smoke emission, special additives are used when CRs containing halogens are being introduced. The most active of these additives are the oxides of aluminium, zinc and tin (Cusack P.A., *Fire and Mater.*, 1986, Vol. 1, No. 1, pp. 41-46).

- 5 The problems with using CR containing halogens are the low resistance of the materials produced to the effect of ultraviolet radiation, their high toxicity and corrosion of the equipment during processing.

The above-listed faults are largely inapplicable to CRs containing phosphorus – Bisphenol-S (Horrocks A.P., *Polym. Degrad. Stab.*, 1996, Vol. 54, pp. 143-154).

- 10 The commercial firm Albright and Wilson market a cyclic phosphonate called Amgard 1045 (Application 2250291 Great Britain MPC C08K 8/03, 7/04).

- The introduction of red phosphorus (1-15 wt.%) and melamicyanurate (4-15 wt.%) into a polyester makes it possible to produce a high-strength material (Application 2250291 Great Britain MPC C08K 8/03, 7/04). However, the process
15 of the application of high-fire-risk red phosphorus is quite complex. Also, the polyester compositions produced acquire a certain coloration.

- The firm "Hoechst" (Germany) produces fireproofed polyester fibres using a bifunctional compound containing phosphorus as a CR. This compound is marketed as Trevira FR and CS (Baranova T.L., Smirnova T.V., Ayzenshteyn E.M.
20 Fireproofed Polyester Fibres. Information Review, Series Chemical Fibre Industry. Moscow, NIITEKhim, 1986, 42 pp.). However, the fireproofing characteristics of these fibres are not high enough, and for a phosphorus content of 0.8-1.0%, the OI is 26-27%.

- One tendency under intensive development in recent years is the introduction
25 of antipyrene additives to polymer compositions in the form of microcapsules.

- Encapsulation methods have been worked out for tetrafluorodibromethane (boiling point 47.5°C) and tetrachlorodifluoroethane (boiling point 92.8°C). Gelatine and gum arabic are used for the shell. The Italian firm "Eurand" has organised the industrial production of microencapsulated tetrafluorodibromethane (freon – 114 B2)
30 (Aleksandrov L.V., Smirnova T.V., Khalturinskiy N.A., *Fireproofed Materials*, Moscow, VNIPI, 1991, 89 pp.).

There are known fireproofing compositions in which the antipyrène is enclosed in a polymer shell, e.g. a composition based on polyolefins, containing as combustion retardant $\text{Al}(\text{OH})_3$ microencapsulated in a polyurethane shell (EP A 04114971 B 1, C 08 K 9/08, 1995), or a composition including microencapsulated tris-(2,3-dibromopropyl)phosphate in a shell of polyvinyl alcohol or urea-formaldehyde resin (US 3660821, cl. 260-2,5, 1972).

Another problem with the known polymer compositions which have a microencapsulated combustion retardant is their high degree of filling with combustion retardant (up to 60%), as a result of which their physical and mechanical indicators are low.

Yet another major problem with the known compositions is the impossibility of processing them at $T > 200^\circ\text{C}$ (i.e., they cannot be moulded), since $\text{Al}(\text{OH})_3$ becomes degraded at $T > 180^\circ\text{C}$, and the polymer shells of the microencapsulated combustion retardants in the known compositions begin to break down even at 160-190°C, leading to the release of the antipyrène from the shell and its decomposition, thus reducing the fire resistance of the compositions and making them more difficult to process.

There is a known polymer composition based on polyolefins, including red phosphorus microencapsulated in melamine formaldehyde resin (EP A 0250662, MPC C 08 K 9/10, 1986). Melamine formaldehyde resin is somewhat more stable than the antipyrène shell in the other known compositions, but at $T > 200-220^\circ\text{C}$, it too begins to decompose, followed by the hydrolysis of the red phosphorus and the formation of highly toxic phosphines. Consequently, this is also a composition which cannot be processed by moulding, since this takes place at temperatures which are too high (250-280°C).

Substance of the invention

In spite of the large number of proposed processes for reducing the combustibility of polymer materials, the problem of creating combustion retardants for polymer materials and more efficient means of producing low-combustibility

polymer compositions remains urgent. This invention is primarily directed towards solving it.

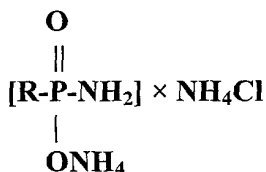
Other problems tackled by the invention are:

- reducing smoke-forming capacity during the pyrolysis and combustion of fireproofed polymer compositions;
- improving the workability of polymer compositions;
- making it possible to implement the developed processes using equipment already installed in production lines for the treatment of polyolefins and synthetic rubbers.

The authors of this invention have previously proposed the use of the microencapsulated antipyrene T-2 as a CR for polyethylene and polypropylene (Zubkova N.S, et al., Plastmassy, 1996, No. 5, pp. 35-36). This is a technical mixture of two individual compounds – the ammonium salt of methylphosphonic acid and ammonium chloride.

The authors later discovered, to their surprise, that a complex compound of the ammonia salt of the amide of methylphosphonic acid with ammonium chloride provides more effective fireproofing than the technical mixture referred to above. In the absence of a theory to explain the reason for this unexpected result, it may be supposed that complex compounds are more active catalysts of the coke-formation processes which are responsible for reducing the combustibility of polymer materials.

Thus, the substance of this invention is primarily the creation of a new combustion retardant for polymer compositions, for which we propose complex compounds of the ammonia salt of the amide of alkylphosphonic acid with ammonium chloride (I)



where R is the alkyl radical C – 1-3.

It was established experimentally that in the said complex compound, there are about 1.8 molecules of ammonium chloride for one molecule of the ammonium salt of the amide of alkylphosphonic acid.

The new complex compound as in Formula (I) can be produced by the interaction of the dichloroanhydride of alkylphosphonic acid with gaseous ammonia in a medium of organic solvent at a temperature of 10-20°C.

The combustion retardant which is the subject of this invention can be used by various methods.

To impart enhanced fireproofing properties to such polymers as polyethylene, polypropylene and the copolymers of various compositions based on them, the created combustion retardant should be introduced at the polymer processing stage.

Thus, for example, the new combustion retardant can be jointly extruded with the polymer, after which the polymer fibre can be moulded and reprocessed into granules.

Another applied-for process for producing polymer materials of the above type is the mixing of the new combustion retardant with polymer composition and then rolling the mass and pressing it into articles.

For the processes described above, and others, for the production of low fire risk polymer materials, when the combustion retardant developed by the authors is introduced into the polymer in the course of its processing, it is advisable first to microencapsulate the combustion retardant in a polymer shell, capsule size being from 5 to 25 µm. To produce the microcapsule shell, one may use polyethylene or polyorganosiloxanes, in particular polyvinylmethyldiethoxysiloxane or polyamino-propylethoxysiloxane. To produce low fire risk polymer materials such as polyester and epoxy resins, the new combustion retardant must be introduced into the polymer composition before it sets.

These compositions may find wide application as binders for glass plastics, sealants, cast insulation and adhesives, as protective coatings for various materials and to produce items by casting in many fields of technology, such as the electrotechnical and electronic fields, and also in construction, aviation, shipbuilding etc.

When set, the compositions produced are solid infusible materials which do not dissolve in organic solvents, are resistant to the effect of acids and alkalis, which have good thermal, physical, mechanical and electrical insulation properties, with no volatile components and which are extinguished on being carried out of a flame.

5 The new combustion retardant can also be used for producing low fire risk synthetic rubbers.

The invention is further illustrated by examples of its implementation.

In these examples:

- the oxygen index, OI, is the minimum content of oxygen in a mixture with nitrogen at which stable combustion of a specimen is maintained after the removal of the source of ignition;
- residual combustion time is the combustion time of the specimen after the removal of the source of ignition;
- fire resistance class PV is a grading from 0 to 4, which was determined in accordance with GOST 28157-89, a state standard of the former USSR.

Examples of the implementation of the invention

Example 1. Production of the complex compound.

300 ml of chloroform are saturated with gaseous ammonia at temperature 10°C. A solution of dichloroanhydride of methylphosphonic acid in chloroform (60 g of dichloroanhydride of methylphosphonic acid dissolved in 200 ml of chloroform) is slowly added, over a period of two hours, to the solution obtained. Ammonia is continuously bubbled through the reaction mixture to maintain the alkaline medium (pH = 9). The temperature of the process should not exceed 20°C. The sediment which forms is filtered off on a Büchner funnel and dried in a vacuum cupboard. The output of the synthesised product is 78.9%. The gross formula is $\text{CH}_{16,3}\text{PN}_{3,8}\text{O}_2\text{Cl}_{1,8}$.

Elemental analysis, found: C 5.8, H 8.1, P 14.3, N 24.9, Cl 30.7; calculated: C 5.8, H 7.8, P 14.9, N 25.5, Cl 30.6.

The formation of the complex compound was proved by the methods of thermogravimetric analysis (TGA), differential-scanning calorimetry (DSC) and X-ray photoelectronic spectroscopy (RPES).

The TGA curve of the complex compound of the ammonia salt of the amide of metaphosphonic acid and ammonium chloride includes one thermo-oxidising decomposition peak in the temperature interval 240-400°C with a maximum at temperature 348°C, which is characteristic of the individual compound. The DSC data show that the synthesised product melts at 202°C (one peak), which is considerably higher than the melting point of the pure ammonia salt of the amide of methylphosphonic acid (124°C).

The RPES spectrum of the synthesised product shows unusually low bond energy of the 2p electrons of the chlorine level (198.1 eV), which indicates the formation of the complex compound. The N1s spectrum includes two main peaks – at bond energy 400.2 eV, corresponding to the P-N bonds, and at bond energy 401.7 eV, corresponding to nitrogen in the form of ammonia, which is considerably lower than the nitrogen bond in NH₄Cl.

Example 2. A composition including 75 g of polypropylene crumbs and 25 g of CR in accordance with this invention is fed into a screw extruder. Moulding takes place at 170°C. The homogeneous melt enters a water bath (18-25°C) and goes for granulation. The modified polyethylene has OI 27.6%, no residual combustion time, fire resistance class PV-0 in accordance with the USSR state standard (GOST 28157-89).

Example 3.

A composition including 75 g of polypropylene crumbs and 25 g of CR in accordance with this invention encapsulated in a polyethylene shell (shell contains 10 wt.% CR, size of microcapsules 25µm) is processed in accordance with Example 1. Moulding temperature – 230°C. The modified polypropylene has OI 28.2%, no residual combustion time, fire resistance class PV-0.

Example 4.

A composition including 90 g polyester crumbs and 10 g CR in accordance with this invention microencapsulated in a shell (shell contains 5 wt.% CR, microcapsule size 10 μ m), is processed in accordance with Example 1. Moulding temperature – 270°C.

- 5 The modified polyester has OI 29.6%, no residual combustion time, fire resistance class – PV-0.

Example 5.

A composition including 85 g polyester crumbs and 15 g CR in accordance with this invention microencapsulated in an ethylene shell (shell contains 2 wt.% CR, microcapsule size 10 μ m), is processed in accordance with Example 1. Moulding temperature – 270°C. The modified polyester has IO 31.0%, no residual combustion time, fire resistance class – PV-0.

15 **Example 6.**

100 g of epoxy resin are mixed with 10 g hardener and 15 g CR in accordance with this invention and allowed to set at room temperature for 48 hours; the solid composition modified in this way becomes a low combustibility material. OI (oxygen index) is 35, no residual combustion time, fire resistance class PV-0.

20

Example 7.

Glass fibre is saturated with an epoxy composition produced in accordance with Example 5 and allowed to set at temperature 60-80°C for 20-30 minutes. The composition obtained contains 40 wt.% binder (epoxy composition) and 60 wt.% filler (glass fibre). The composition material is of low combustibility, no residual combustion time, fire resistance class PV-0.

25

Example 8.

A composition consisting of 60 g unsaturated polyester resin, 15 g CR in accordance with this invention microencapsulated in a polyaminopropylethoxysiloxane shell (shell contains 5 wt.% CR, microcapsule size 15 μm) and 25 g of staple fibre (viscose, polycaproatamide) was pressed at temperature 180°C and pressure 80 kg/cm². The plastics produced have OI 29.5%, no residual combustion time.

Example 9.

A composition consisting of 80 g rubber mixture including butadiene styrene rubber and 20 g CR in accordance with this invention is thoroughly mixed, rolled at temperature 140-150°C and the articles are then pressed at temperature 170-180°C. The modified rubber mixture has OI 28%, no residual combustion time.

Example 10.

A composition consisting of 85 g rubber mixture based on isoprene rubber and 15 g CR in accordance with this invention microencapsulated in a polyaminopropylethoxysiloxane shell (shell contains 5 wt.% CR, microcapsule size 15 μm) is processed in accordance with Example 5. The modified rubber composition has OI 28.1%, no residual combustion time.

Example 11.

A composition consisting of 80 g polymethyl methacrylate and 20 g CR in accordance with this invention is processed in accordance with Example 1 at temperature 220°C. The modified polymethyl methacrylate has OI 27.2%, no residual combustion time.

Example 12.

A composition consisting of 75 g polycaproatamide (PCA) and 25 g CR in accordance with this invention microencapsulated in an ethylene shell (shell contains 10 wt.% CR, microcapsule size 25 μm) is processed in accordance with Example 1. Moulding

temperature 230°C. The modified PCA has OI 29%, no residual combustion time, fire resistance class PV-0.

Example 13 (comparative)

- 5 A composition including 85 g polyester crumbs and 15 g technical mixture consisting of 7.7 g ammonia salt of amide of methylphosphonic acid and 7.3 g ammonium chloride is processed in accordance with Example 4. The modified polyester has OI 27.6%.

10 **Example 14** (comparative)

A composition including 75 g polypropylene crumbs and 25 g of a technical mixture including 12.8 g ammonia salt of amide of methylphosphonic acid and 12.2 g ammonium chloride is processed in accordance with Example 3. The modified polyester has OI 24.8%, fire resistance class PV-1.

15

Example 15 (comparative)

A composition including 75 g polyethylene crumbs and 25 g of a technical mixture including 12.8 g ammonia salt of amide of methylphosphonic acid and 12.2 g ammonium chloride is processed in accordance with Example 2. The modified polyester has OI 24.8%, fire resistance class PV-1.

20

Example 16 (comparative)

A composition including 75 g polycaproamide crumbs and 25 g of a technical mixture including 12.8 g ammonia salt of amide of methylphosphonic acid and 12.2 g ammonium chloride is processed in accordance with Example 12. The modified polyester has OI 24.8%, fire resistance class PV-1.

25

The comparative examples illustrate the fact that the proposed complex compound is a more effective antipyrene for polyethylene (Examples 2-15)
30 polypropylene (Examples 3-14), polyester (Examples 5-13) and other polymers than

a technical mixture of the two individual compounds of ammonia salt of diamide of methylphosphonic acid and ammonium chloride.

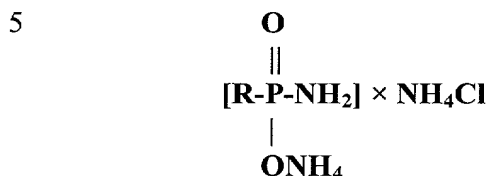
Furthermore, the diamide has low resistance to the action of hydrolising agents, and even under the effect of moisture in the air, diamide of methylphosphonic acid gradually passes through the ammonia salt into the methylphosphonic acid.

Therefore, the use of the proposed complex is a qualitatively new solution to the problem of reducing the combustibility of polymer materials.

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CLAIMS:

1. Complex compound of ammonia salt of amide of alkylphosphonic acid with ammonium chloride of formula (I)



10 where R is the alkyl radical C-1-3.

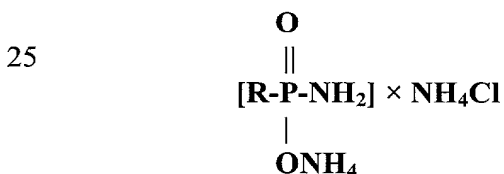
2. Complex compound in accordance with Claim 1, in which there are about 1.8 molecules of ammonium chloride to one molecule of ammonia salt of amide of alkylphosphonic acid.

15

3. Process for producing a complex compound of ammonia salt of amide of alkylphosphonic acid with ammonium chloride of Formula (I), consisting of the interaction of dichloroanhydride of alkylphosphonic acid with gaseous ammonia in a medium of organic solvent at temperature 10-20°C.

20

4. Combustion retardant for polymer materials, consisting of a complex compound of ammonia salt of amide of alkylphosphonic acid with ammonium chloride of formula (I)



where R is the alkyl radical C-1-3.

30

5. Combustion retardant in accordance with Claim 4, in which there are about 1.8 molecules of ammonium chloride to one molecule of ammonia salt of amide of alkylphosphonic acid.

6. Combustion retardant in accordance with Claim 4 or 5, *characterised* in that it is microencapsulated in a polymer shell.

7. Combustion retardant in accordance with Claim 6, *characterised* in that the said
5 polymer shell is made of polyethylene.

8. Combustion retardant in accordance with Claim 6, *characterised* in that the said polymer shell is made of polyorganosiloxanes.

10 9. Combustion retardant in accordance with Claim 8, *characterised* in that the polyorganosiloxanes are selected from a group including polyvinylmethyldiethoxysiloxane and polyaminopropylethoxysiloxane.

15 10. Process for producing low fire risk polymer materials by the introduction of the CR into the polymer in the course of its processing, *characterised* in that the CR used is a complex compound of ammonia salt of amide of alkylphosphonic acid with ammonium chloride of Formula (I).

20 11. Process for producing low fire risk polymer materials in accordance with Claim 10, *characterised* in that it includes the following sequence of operations:
- joint extrusion of the said combustion retardant with the polymer;
- moulding the polymer fibre;
- granulation.

25 12. Process for producing low fire risk polymer materials in accordance with Claim 10, *characterised* in that it includes the following sequence of operations:
- mixing of the said combustion retardant with the polymer composition;
- rolling the mass;
- pressing the articles.

30

13. Process in accordance with any of Claims 10-12, *characterised* in that the combustion retardant is first microencapsulated in a polymer shell.

14. Process in accordance with Claim 13, *characterised* in that the size of the microcapsules is 5-25 μm .

15. Process in accordance with Claim 13, *characterised* in that the polymer shell is made of polyethylene with shell content including 10-15 wt.% of combustion retardant.

16. Process in accordance with Claim 13, *characterised* in that polyorganosiloxanes are used for the polymer shell.

17. Process in accordance with Claim 16, *characterised* in that the polyorganosiloxane consists of polymethyldiethoxysiloxanes with shell containing 2-5 wt.% of combustion retardant.

18. Process in accordance with Claim 16, *characterised* in that the polyorganosiloxane used is polyaminopropylethoxysiloxane, with shell containing 2-5 wt.% of combustion retardant.

19. Process in accordance with any of Claims 10-18, *characterised* in that polyethylene, polypropylene and copolymers of various compositions based on them are processed.

20. Process in accordance with any of Claims 10-18, *characterised* in that polystyrene and copolymers of various compositions based on it are processed.

21. Process for producing low fire risk polymer materials by the introduction of combustion retardant into the polymer, *characterised* in that the combustion retardant used is a complex compound of ammonia salt of amide of alkylphosphonic acid with

ammonium chloride of Formula (I), which is introduced into the polymer composition before it sets.

22. Process in accordance with Claim 21, *characterised* in that a filler is introduced
5 into the polymer composition along with the said combustion retardant, and as a result of the saturation of the filler with the setting polymer composition, low fire risk materials are produced.

23. Process in accordance with Claim 21 or 22, *characterised* in that polyesters are
10 processed.

24. Process in accordance with Claim 21 or 22, *characterised* in that epoxy resins are processed.

25. Process for producing low fire risk polymer materials, *characterised* in that a complex compound of ammonia salt of amide of alkylphosphonic acid with ammonium chloride of Formula (I) is introduced into a polymer composition including synthetic rubber, after which it is rolled before the article is pressed.

26. Low fire risk polyethylene produced by a process in accordance with Claim 6.

27. Low fire risk polypropylene produced by a process in accordance with Claim 6.

28. Low fire risk polystyrene produced by a process in accordance with Claim 6.

29. Low fire risk copolymers based on polyethylene, polypropylene and polystyrene, produced by a process in accordance with Claim 6.

30. Low fire risk polyesters produced by a process in accordance with Claim 21.

31. Low fire risk epoxy resins produced by a process in accordance with Claim 21.

32. Low fire risk composition materials produced by a process in accordance with Claim 22.

5 33. Low fire risk synthetic rubbers produced by a process in accordance with Claim 25.

34. Low fire risk polycaproamide materials produced by a process in accordance with Claim 13.

10

35. Low fire risk polymethyl methacrylate compositions produced by a process in accordance with Claim 11.

ABSTRACT

A description is given of a combustion retardant for polymer materials in the form of a new complex compound of the ammonium salt of amide of
5 alkylphosphonic acid with ammonium chloride, and also of processes for producing various polymer materials using the said combustion retardant.

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| Filing Date | 06 March 2001 |
| First Named Inventor | Zubkova |
| Title | "Combustion Retardant." |
| Group Art Unit | |
| Examiner Name | |
| Attorney Docket Number | A01058US (98148.13) |

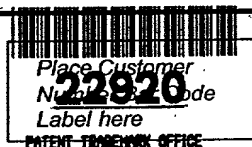
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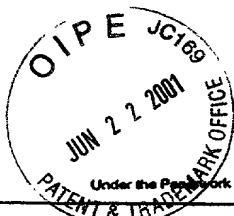
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Я, нижеупомянутый изобретатель, настоящим подтверждаю, что:

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Я убежден, что я являюсь первоначальным, первым и единственным изобретателем (если ниже указано только одно имя), или одним из первоначальных и первых со-авторов (если ниже указаны несколько имен) заявляемого изобретения, на которое запрашивается патент и которое называется:

Описание изобретения приложено к сему (если в расположенной ниже клетке нет отметки):

☐ было подано /дата/ _____
как заявка США номер или международный
PCT № _____
с изменениями, внесенными /дата/ _____
(если требуется).

Настоящим я заявляю, что я изучил и понимаю содержание вышеназванного описания, включая формулу изобретения со всеми поправками, указанными выше.

Я признаю обязанность сообщить информацию, необходимую для патентования в соответствии с §1.56 раздела 37 Кодекса Федеральных Правил.

Настоящим я предъявляю иностранные преимущественные права приоритета в соответствии с §119 (a)-(d) или §365 (b) раздела 35 Кодекса Соединенных Штатов на любую(ые) иностранную(ые) заявку(и) на патент или авторское свидетельство, или с §365 (a) на любую международную заявку PCT, назначившую одну или больше стран кроме Соединенных Штатов, перечисленную(ые) ниже, а также указал ниже с расположением отметки в клетке все иностранные заявки на патент или авторское свидетельство или международную заявку PCT, поданные ранее, чем заявка, на которую предъявлено притязание на приоритет.

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated next to my name.

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled

COMBUSTION RETARDANT FOR
POLYMERIC MATERIALS

the specification of which is attached hereto unless the following box is checked:

☒ was filed on 2 AUGUST 1999
as United States Application Number or PCT
International Application Number PCT/RU99/00273
and was amended on _____
(if applicable).

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations, § 1.56.

I hereby claim foreign priority under Title 35, United States Code, § 119(a)-(d) or § 365(b) of any foreign application(s) for patent or inventor's certificate, or § 365(a) of any PCT International application which designated at least one country other than the United States, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate, or PCT International application having a filing date before that of the application on which priority is claimed.

Prior Foreign Application(s)
Презняя(ие) иностранная(ые) заявка(и)

Притязание на приоритет не предъявляется

Priority Not Claimed

Номер (Number) Страна (Country)

День/Месяц/Год подачи
(Day/Month/Year Filed)

☐

PCT/RU98/00289

RUSSIA

08 SEPTEMBER 1998

☐

Номер (Number) Страна (Country)

День/Месяц/Год подачи
(Day/Month/Year Filed)

Номер (Number) Страна (Country)

День/Месяц/Год подачи
(Day/Month/Year Filed)

☐

Настоящим я предъявляю иностранные
преимущественные права приоритета в
соответствии с § 119 (e) раздела 35 Кодекса
Соединенных Штатов на любую(ые)
предварительную(ые) заявку(и), перечисленную(ые)
ниже.

I hereby claim the benefit under Title 35, United States Code,
§ 119(e) of any United States provisional application(s) listed
below.

(Заявка №)

(Дата подачи заявки)

(Application No.)

(Filing Date)

(Заявка №)

(Дата подачи заявки)

(Application No.)

(Filing Date)

Настоящим я заявляю претензию на выгоду, в
соответствии с § 120 раздела 35 Кодекса
Соединенных Штатов, от всех нижеперечисленных
заявок(ки) США или с § 365 (c) от любой
международной заявки PCT, назначившей
Соединенные Штаты, в той мере, в которой предмет
изобретения в каждом пункте, на который заявлен
приоритет, не был раскрыт в поданной ранее заявке
США или международной заявке PCT, как это
предусмотрено в первом абзаце § 112 раздела 35
Кодекса Соединенных Штатов. Я признаю
обязанность раскрыть информацию, которая
является вещественной для патентоспособности, как
это предусмотрено в § 1.56 раздела 37 Кодекса
Федеральных Правил, которая стала доступна за
период времени между подачей предшествующей
заявки и датой подачи национальной или
международной заявки PCT.

I hereby claim the benefit under Title 35, United States Code,
§ 120 of any United States application(s), or § 365(c) of any PCT
international application designating the United States, listed
below and, insofar as the subject matter of each of the claims of
this application is not disclosed in the prior United States or PCT
international application in the manner provided by the first
paragraph of Title 35, United States Code, § 112, I acknowledge
the duty to disclose information which is material to patentability
as defined in Title 37, Code of Federal Regulations, § 1.56 which
became available between the filing date of the prior application
and the national or PCT international filing date of this application.

Under the Paperwork Reduction Act of 1996, no persons are required to respond to a collection of information unless it displays a valid OMB control number.

(Заявка №)

(Дата подачи заявки)

(Статус - запатентовано, рассматривается, заявитель отказался)

(Application No.)

(Filing Date)

(Status - patented, pending, abandoned)

(Заявка №)

(Дата подачи заявки)

(Статус - запатентовано, рассматривается, заявитель отказался)

(Application No.)

(Filing Date)

(Status - patented, pending, abandoned)

Настоящим подтверждаю, что все заявления, сделанные здесь на основе моих знаний, являются правдой, и я также верю в достоверность всех заявлений, основанных на доступной мне информации и убеждениях; кроме того, эти заявления были сделаны со знанием того, что умышленно ложные заявления и подобные им действия караются штрафом, или тюремным заключением, или тем и другим, в соответствии со статьей 1001 раздела 18 Кодекса Соединенных Штатов, и что такие ложные сведения могут сделать недействительной как эту заявку, так и любой патент, по ней выданный.

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

1 - 00 Nina Sergeevna ZUBKOVA
Полное имя единственного или первого автора Full name of sole or first inventor
изобретения

Подпись автора изобретения Дата Inventor's signature Date
x *Nina* x 23.05.2001

Местожительство Residence
Moscow, Russia RUx

Гражданство Citizenship
Russia

Почтовый адрес Post Office Address
Russia 117574 Moscow, Novoyasenevsky
prospekt, d.22, korp.1, kv.592

2 - 00 Nataliya Grigorievna BUTYLKINA
Полное имя второго автора изобретения Full name of second joint inventor, if any
(если имеется)

Подпись автора изобретения Дата Second inventor's signature Date
x *Nataliya* x 23.05.2001

Местожительство Residence
Moscow, Russia RUx

Гражданство Citizenship
Russia

Почтовый адрес Post Office Address
Russia 113461 Moscow, ul. Kakhovka,
d.14, korp.2, kv.103

(Аналогичная информация о третьем и последующих
авторах изобретения должна быть представлена, а
также их подписи)

(Supply information and signature for third and subsequent joint
inventors.)

3 - 00 Nikolai Alexandrovich KHALTURINSKY
Полное имя единственного или первого автора изобретения Full name of sole or first inventor

Подпись автора изобретения Дата Inventor's signature Date
[Signature] *23.05.2001*

Местожительство Residence
Moscow, Russia R U X

Гражданство Citizenship
Russia

Почтовый адрес Post Office Address
Russia 117229 Moscow, ul.

Ulyanova, d.12, korp.2, kv.17

4 - 00 Alexandr Alexandrovich BERLIN
Полное имя второго автора изобретения Full name of second joint inventor, if any
(если имеется)

Подпись автора изобретения Дата Second inventor's signature Date
[Signature] *23.05.2001* *[Signature]* *23.05.2001*

Местожительство Residence
Moscow, Russia R U X

Гражданство Citizenship
Russia

Почтовый адрес Post Office Address
Russia 117419 Moscow, ul.

Donskaya, d.24, kv.68

(Аналогичная информация о третьем и последующих авторах изобретения должна быть представлена, а также их подписи)

(Supply information and signature for third and subsequent joint inventors.)

Applicant or Patentee. Howard M. Chandler & Marc Sinatra Attorney's Dkt. No. 0141-2006
Serial or Patent No 09/856,105
Filed or Issued: May 17, 2001
For: A METHOD OF DETECTING THE PRESENCE OF AN ANALYTE IN A BIOLOGICAL SAMPLE

**VERIFIED STATEMENT (DECLARATION) CLAIMING SMALL ENTITY
STATUS [37 19(f) and 1.27(c)] - SMALL BUSINESS CONCERN**

I hereby declare that I am

- ☐ the owner of the small business concern identified below:
☒ an official of the small business concern empowered to act on behalf of the concern identified below:

NAME OF CONCERN ENTERIX INC. of 348 US Route One, Falmouth,
ADDRESS OF CONCERN Maine 04105, United States of America

I hereby declare that the above identified small business concern qualifies as a small business concern as defined in 13 CFR 121.3-18, and reproduced in 37 CFR 1.9(d), for purposes of paying reduced fees under section 41(a) and (b) of Title 35, United States Code, in that the number of employees of the concern, including those of its affiliates, does not exceed 500 persons. For purposes of this statement (1) the number of employees of the business concern is the average over the previous fiscal year of the concern of the persons employed on a full-time, part-time or temporary basis during each of the pay periods of the fiscal year, and (2) concerns are affiliates of each other when either, directly or indirectly, one concern controls or has the power to control the other, or a third party or parties controls or has the power to control both.

I hereby declare that rights under contract or law have been conveyed to and remain with the small business concern identified above with regard to the invention, entitled:

A METHOD OF DETECTING THE PRESENCE OF AN ANALYTE IN A BIOLOGICAL SAMPLE

by inventors Howard Milne CHANDLER and Marc SINATRA described in
the specification filed herewith. International Patent Application No PCT/AU99/01014 filed
application Serial No. _____, filed 17 November, 1999
patent No. _____, issued _____

If the rights held by the above-identified small business concern are not exclusive, each individual, concern or organization having rights to the invention is listed below* and no rights to the invention are held by any person, other than the inventor who could not qualify as an independent inventor under 37 CFR 1.9(c) if that person made the invention, or by any concern which would not qualify as a small business concern under 37 CFR 1.9(d), or a nonprofit organization under 37 CFR 1.9(e). *NOTE Separate verified statements are required from each named person, concern or organization having rights to the invention averring to their status as small entities (37 CFR 1.27)

Name _____

Address _____

☐ Individual ☐ Small Business Concern ☐ Nonprofit Organization

Name _____

Address _____

☐ Individual ☐ Small Business Concern ☐ Nonprofit Organization

I acknowledge the duty to file, in this application of patent, notification of any change in status resulting in loss of entitlement to small entity status prior to paying, or at the time of paying, the earliest of the issue fee or any maintenance fee due after the date on which status as a small entity is no longer appropriate. [37 CFR 1.28(b)]

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under § 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application, any patent issuing thereon, or any patent to which this verified statement is directed

NAME OF PERSON SIGNING Howard Milne Chandler

TITLE OF PERSON OTHER THAN OWNER CEO

ADDRESS OF PERSON SIGNING 857 Prince's Point Rd, Yarmouth, Maine 04096

SIGNATURE H.M. Chandler

DATE May 25, 2001